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OPTICAL AND OTHER PHYSICAL PROPERTIES OF $\text{Al}(\text{PO}_3)_3$ - CONTAINING FLUORO-PHOSPHATE GLASSES

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Résumé. - Dans cet article, on étudie les propriétés optiques et d'autres propriétés physiques de verre fluorophosphate comprenant le système à deux composantes de $\text{Al}(\text{PO}_3)_3$ et l'influence de divers fluorures sur ces propriétés dans le verre de basse teneur de $\text{Al}(\text{PO}_3)_3$. On donne le site de verre de basse teneur de $\text{Al}(\text{PO}_3)_3$ à la figure de n_d-v , et une partie des propriétés de divers fluorures dans le verre est calculée.

Abstract. - In this paper, the optical and some other physical properties of fluorophosphate glasses in binary systems containing $\text{Al}(\text{PO}_3)_3$ and the influence of various fluorides on the properties of glass with low $\text{Al}(\text{PO}_3)_3$ content have been studied. The positions of low $\text{Al}(\text{PO}_3)_3$ content glasses in n_d-v diagram have been plotted and the partial properties of fluorides in glass have been calculated.

Fluorophosphate glass is a glass with special optical properties such as high Abbe value, transmittance in near UV and IR regions and low nonlinear refractive index (n_2), it is now under development. Comparing the properties of various glassy metaphosphate, it shows that $\text{Al}(\text{PO}_3)_3$ possesses a higher refractive index, a greater Abbe value^[1] as well as better physical and chemical properties. Therefore $\text{Al}(\text{PO}_3)_3$ has been introduced into many fluorophosphate glasses. The authors have already studied the formation and structure of $\text{Al}(\text{PO}_3)_3$ - containing fluorophosphate glasses^[2], and in the present work, the optical and some other physical properties of those glasses are investigated with the emphasis placed on the low $\text{Al}(\text{PO}_3)_3$ content glasses.

The following glass systems were selected for the investigation

- 1) $(100-x) \text{Al}(\text{PO}_3)_3 \cdot x \text{RF}_n$ (R = Li, Na, Ba, Sr, Al).
- 2) $(10 \text{Al}(\text{PO}_3)_3 \cdot 20 \text{AlF}_3 \cdot 50 \text{SrF}_2 \cdot 20 \text{LiF}) + 5 \text{RF}_n$ (R = Li, Na, K, Be, Mg, Ca, Sr, Ba, Zn, Cd, Pb, Al, Gd).
- 3) $(5 \text{Al}(\text{PO}_3)_3 \cdot 28 \text{AlF}_3 \cdot 30 \text{SrF}_2 \cdot 27 \text{CaF}_2 \cdot 10 \text{LiF}) + 3 \text{RF}_n$ (R = Li, Na, K, Mg, Ca, Sr, Ba, Zn, Al, Ga).

Aluminium metaphosphate and fluorides were used as starting materials. Glasses with high $\text{Al}(\text{PO}_3)_3$ content were melted in sintered alumina crucibles, and others in platinum crucibles. Different melting conditions were selected according to different glass systems.

The refractive indices (n_d , n_F , n_C , n_g), the thermal expansion coefficients α , the moduli of elasticity E , and the densities of the glasses were determined, the molecular volumes of glasses, the average atomic volumes of fluorine and oxygen V_{OF} , and the partial refractive indices and Abbe values of various fluorides in glasses were calculated.

1) In the previous work^[2], on the basis of experimental results of glass formation, devitrification and structure study, we pointed out that the network of the glasses with low $\text{Al}(\text{PO}_3)_3$ content is greatly destroyed. Therefore the property variation of these glasses should be similar to that of the oxide glasses in the destroyed region. In general, the refractive index and dispersion increase with increasing ionic radius r , while α and E vary with the electrostatic force Z/a^2 between cations and fluorine ions (Fig.1 and 2).

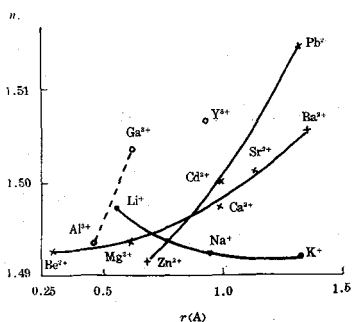


Fig.1 n_d vs $r(\text{Å})$ in the system with 10 mol% $\text{Al}(\text{PO}_3)_3$

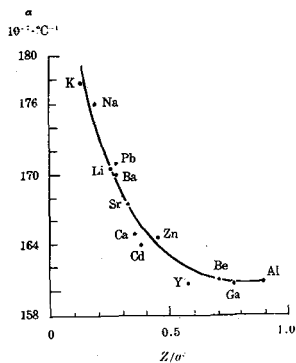


Fig.2 α_{30-Tg} vs Z/a^2 in the system with 10 mol% $\text{Al}(\text{PO}_3)_3$

2) In the region with low $\text{Al}(\text{PO}_3)_3$ content, some fluorides such as LiF , MgF_2 and AlF_3 could reconnect the disrupted glass network^[2]. This is also reflected in the variation of the physical properties of glass. In $\text{Al}(\text{PO}_3)_3 - \text{RF}_n$ systems, those containing LiF or AlF_3 possess smaller V_{OF} (Fig.3), and with the increase in the RF_n content, the increase in V_{OF} is relatively slow. The LiF , MgF_2 , AlF_3 -containing glasses all have higher E and lower V_{OF} (Fig.4,5), it can be considered as the increase in compactness of glass structure.

3) In the glasses with very low $\text{Al}(\text{PO}_3)_3$ content, besides the particular role of

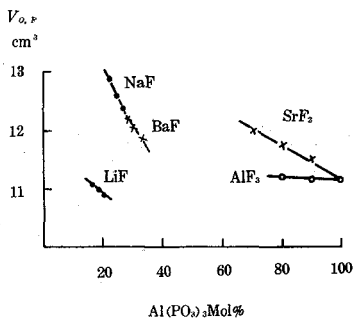


Fig.3 Dependence of V_{0F} on the $Al(PO_3)_3$ content in $Al(PO_3)_3 - RF_n$ systems

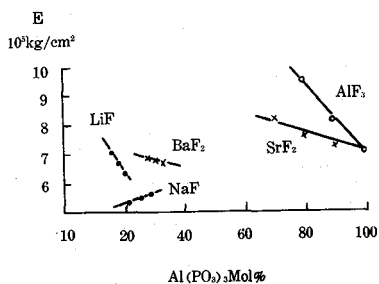


Fig.4 Dependence of E on the $Al(PO_3)_3$ content in $Al(PO_3)_3 - RF_n$ systems

LiF, MgF_2 , AlF_3 mentioned above, other cations filling in the interspace between structural chains can also enhance the glass structure to some extent due to the electrostatic attraction. Generally speaking, V_{0F} depends on the related ionic radii. However, for the glasses with low $Al(PO_3)_3$ content, from the diagram of V_{0F} vs Z/a^2 (Fig.5), it can be obviously seen that V_{0F} decreases with increasing Z/a^2 . Combining this and the variations in α as well as E , it can be considered that electrostatic force has relatively great influence on the properties of glass when the glass network is heavily destroyed.

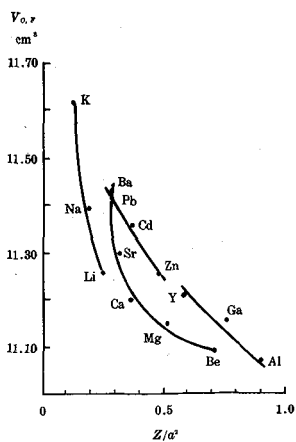


Fig.5 Variation of V_{0F} in systems with 10% $Al(PO_3)_3$ as a function of Z/a^2

4) Fig.6 shows the position of the studied glasses containing 5%, 10% $Al(PO_3)_3$ in $n_d \sim \nu$ diagram with the addition of various fluorides. Using the optical constants of all the glasses studied in this work, the partial refractive indices and Abbe values of some fluorides introduced have been calculated, and the results obtained are listed in Table 1 together with some data from literature. From Table 1, we can see that there is great deviations among the data from different sources. Nevertheless, we can also see that with the increase in the metaphosphate content, the partial refractive index increases, while the ν decreases and the influence of fluorides on the properties of glass lessens. Hence, the partial properties of fluorides can only be suitable for calculating physical properties of glasses in a certain region.

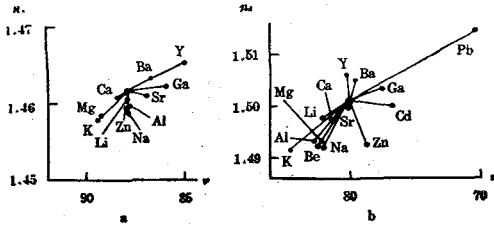


Fig.6 Positions of glasses with addition of fluorides on n_d-v diagram
 a. $Al(PO_3)_3 = 5 \text{ mol\%}$
 b. $Al(PO_3)_3 = 10 \text{ mol\%}$

Table 1. The Partial Properties of Some Fluorides

Source	Our work		Ref. [1]		Ref. [3]		Ref. [3]			
	$Al(PO_3)_3 < 10$	$Al(PO_3)_3 > 30$	$Al(PO_3)_3 < 15$	$Al(PO_3)_3 \sim 30$	-	-	-	-		
Fluoride	n_d	v	n_d	v	n_d	v	n_d	v		
LiF	1.43	110	1.549	-	1.45	109	1.479	78	1.494	-
NaF	1.314	121	-	-	1.37	118	-	-	-	-
MgF ₂	1.321	121	1.47	-	1.41	133	1.461	86	1.47	76
CaF ₂	1.413	104	1.516	-	1.47	81	1.465	80	1.483	76
SrF ₂	1.455	84	1.53	62	1.531	70.8	1.471	79	1.50	75
BaF ₂	1.567	74	1.607	-	1.59	63.4	-	-	-	-
AlF ₃	1.333	120	1.486	78.6	1.40	128	1.424	98	1.40	101

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